Driven Rydberg atoms reveal quartic level repulsion

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The dynamics of Rydberg states of a hydrogen atom subject simultaneously to uniform static electric field and two microwave fields with commensurate frequencies is considered in the range of small fields amplitudes. In the certain range of the parameters of the system the classical secular motion of the electronic ellipse reveals chaotic behavior. Quantum mechanically, when the fine structure of the atom is taken into account, the energy level statistics obey predictions appropriate for the symplectic Gaussian random matrix ensemble.

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Quantum chaos considers correlations between the quantal properties of dynamical system and its classical behavior. In particular level statistics of quantum systems chaotic in the classical limit should generically obey Random Matrix Theory (RMT) predictions [1] according to the famous conjecture of Bohigas, Giannoni and Schmit [2]. The link between RMT and chaotic systems has been quite fruitful for quantum chaos studies (see for reviews [3,4]).

Depending on the symmetries of a given strongly chaotic system, statistical properties of its quantum spectrum fall into one of the three classes known from RMT: orthogonal, unitary and symplectic. The orthogonal class is typically associated with Hamiltonians invariant with respect to some generalized time-reversal symmetry (referred to also as an antiunitary symmetry), the corresponding statistical ensemble of random matrices is referred to as Gaussian Orthogonal Ensemble (GOE). In the absence of any such a symmetry the corresponding class is known as Gaussian Unitary Ensemble (GUE). Finally energy levels of half-integer spin systems with no geometrical symmetries and obeying the generalized time-reversal symmetry T squaring to minus unity ($T^2 = -1$) reveal twofold Kramers degeneracy and their statistical properties pertain to Gaussian Symplectic Ensemble (GSE).

The conjecture has been tested on a number of theoretical models (we refer the reader to reviews [3,4] rather than numerous original papers). Experiments in atomic systems have been restricted to the orthogonal universality class [5] only. To break antiunitary symmetries in atomic species one needs, e.g., a static magnetic field, nonhomogeneous on the atomic scale [3] which is quite hard to realize. For that reason GUE type statistics have been observed experimentally for microwave billiards (so-called *wave chaos* experiments) where the time-reversal symmetry could be broken by applying some ferrite devises [6,7].

As shown by us recently [8,9] atomic systems also allow to generate GUE type statistics provided one uses not only the static fields but also microwaves. Then, instead of considering properties of eigenvalues of a given Hamiltonian H (which is not possible for time-dependent microwave perturbation) one considers quasienergies of the Floquet operator $\mathcal{H} = H - i\hbar\partial/\partial t$ well defined for a periodic driving [10]. For appropriately chosen combination of elliptically polarized microwaves and a static electric field, the (generalized) time-reversal symmetries are broken and GUE type statistics may be observed. Even for weak fields when Floquet states may be thought of as the perturbed principal quantum number n_0 hydrogenic manifold, the typical splitting between levels may be of the order of a few MHz making the experiment feasible. A RMT type level statistics is a manifestation of the classically chaotic behavior as observed (via classical perturbation theory) for the motion of the electronic ellipse (the so-called secular motion).

With GOE or GUE type of statistics realized for atomic species it seems natural to ask whether it is possible to observe also statistics corresponding to the third universality class – the symplectic ensemble. The aim of this letter is to provide an example of such a situation. Such a behavior has not yet been observed, as far as we know, neither for atomic nor for billiard type systems.

To observe the effect one needs an interaction explicitly depending on the electron half-integer spin, an interaction sufficiently intense to relatively strongly perturb the levels. For hydrogen atoms two obvious candidates are the Zeeman effect and the spin-orbit coupling. In the former case, however, there is no time-reversal symmetry squaring to minus unity. The latter seems quite weak in the semiclassical Rydberg states regime. For that reason traditionally the spin-orbit coupling is neglected while considering strongly externally perturbed Rydberg states.

It is another story if we restrict ourselves to small ex-

ternal perturbations and consider the combined effect of these perturbations and the spin-orbit coupling. Our previous experience [8,9] still indicates that the secular motion of the electronic ellipse may be strongly chaotic even for weak (but judiciously chosen) combination of the external fields. Then, if also spin-orbit coupling is taken into account, one may hope to observe GSE type statistics.

Thus we consider the very same model as before, i.e., the hydrogen Rydberg atom driven by microwaves and placed in a static uniform electric field and we add the spin-orbit interaction. The configuration of the external fields must be chosen so that: the time-reversal symmetry with $T^2 = -1$ is preserved, any other geometrical symmetry is broken and the dynamics is irregular classically. To fulfill the first condition we have to use microwaves of linear polarization, in contrast to our earlier studies [8,9]. The Hamiltonian of the system in atomic units is

$$H = \frac{\mathbf{p}^2}{2} - \frac{1}{r} + H_1 + \alpha^2 H_2, \qquad (1)$$

where α is the fine structure constant. H_1 is the external perturbation

$$H_1 = \mathbf{E} \cdot \mathbf{r} + Fx \cos \omega t + F'z \cos 2\omega t \qquad (2)$$

while

$$H_2 = -\frac{\mathbf{p}^4}{8} + \frac{1}{2r^3} \mathbf{L} \cdot \mathbf{S} + \frac{\pi}{2} \delta(\mathbf{r}), \qquad (3)$$

gives the lowest order relativistic corrections [11]. Among them the most important for the present contribution is the spin-orbit coupling. In the above formulae $\mathbf{E}$ denotes a static electric field vector, F and F' are amplitudes of two microwave fields with frequencies ω and 2ω and polarized along the Ox and Oz axes, respectively.

The Hamiltonian (1) is time reversal-invariant, with time-reversal operator squaring to minus unity. Thus eigenvalues of the Floquet Hamiltonian must reveal a twofold Kramers degeneracy [3]. The role of F' field is to break the geometrical symmetry which is preserved by (1) if $F' = 0$. Indeed if we choose, for $F' = 0$, the Oz axis such that a static field component parallel to it vanishes, $E_z = 0$, then the parity operator $\Pi_z = \exp(i\pi S_z)P_z$ (where P_z is a reflection with respect to Oxy plane) with the property $\Pi_z^2 = -1$ commutes with H. This unitary symmetry splits the quasienergy spectrum into two blocks with identical eigenvalues (Kramers degeneracy). Each block is in itself a hermitian matrix, i.e. its eigenvalues are expected to obey GUE but not GSE statistics if the underlying classical motion is chaotic [3].

Since both the microwaves and the static electric field are assumed to be weak, the model may be analyzed perturbatively both classically and quantum mechanically. The latter is straightforward in the effective Hamiltonian formalism [12] with the fields taken in the lowest

nonvanishing order (first order for the static electric field due to a linear Stark effect as well as for the spin-orbit interaction and second order in microwave amplitudes F and F'). The resulting matrices of dimension $2n_0^2$ with n_0 being the principal quantum number of the perturbed manifold studied have been diagonalized with standard routines. We refer the reader to [8] for practical details described there for a similar model.
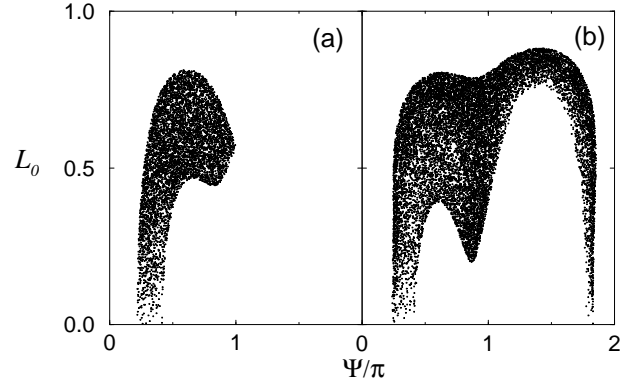

FIG. 1. Poincaré surface of section (at $\Phi = 0$) of the classical secular motion, Eq. (4), for the hydrogen atom in static electric and microwave fields for the energy $(H_0^{\text{eff}} + 1/2)/E_0 = -1$ and with the amplitude and frequency of the microwave field $F_0^2/E_0 = 2$, $\omega_0 = 1.304$, respectively. The coordinates used for the plot are the scaled angular momentum $L_0 = L/n_0$ and its canonically conjugate angle Ψ. Panel (a) corresponds to the orientation of the static electric field vector $\theta = \pi/2$, $\varphi = \pi/4$ while panel (b) is related to the system additionally perturbed by F' field with the amplitude $F_0'^2/E_0 = 5$ (the static field orientation is $\theta = 0.31\pi$, $\varphi = \pi/4$). Note that, for the parameters chosen, not the whole (L_0, Ψ) space is accessible.

In an analogous way one may construct an effective classical Hamiltonian averaging over the phase of the field and the fast motion along the electronic ellipse and considering the slow secular motion of the ellipse itself only. For the classical analysis we neglect the relativistic correction part H_2 (the spin-orbit interaction and Darwin term, $\delta(\mathbf{r})$, do not have direct classical analogs). For the purpose of the present analysis we assume the microwaves to be off resonant thus the perturbation calculations follow the Lie approach of [13,14,9] rather than that appropriate for resonant driving [8].

The resulting classical effective Hamiltonian (first order in E and second order in F and F') takes a form

$$H_0^{\text{eff}} = -\frac{1}{2} + H_{1,0}^{\text{eff}}(\omega_0, F_0^2, F_0'^2, E_0; L_0, \Psi, M_0, \Phi) \quad (4)$$

where $H_0^{\text{eff}} = n_0^2 H^{\text{eff}}$, $F_0 = n_0^4 F$, $F_0' = n_0^4 F'$, $E_0 = n_0^4 E$, $\omega_0 = n_0^3 \omega$, $L_0 = L/n_0$ and $M_0 = M/n_0$ are scaled variables (L, M being the angular momentum and its projection on the Oz axis while Ψ and Φ are angles con-

jugate to L and M, respectively). Observe that classical dynamics depends only on the reduced energy via scaled variables (no dependence on n_0). Fig. 1a shows an example of Poincaré surface of section obtained for $(H_0^{\text{eff}} + 1/2)/E_0 = -1$ with $F_0' = 0$, $F_0^2/E_0 = 2$, $\omega_0 = 1.304$ and orientation of the static field vector $\theta = \pi/2$, $\varphi = \pi/4$ (where θ, φ are usual spherical angles). In Fig. 1b there is a similar plot but for the case with broken the parity symmetry, i.e. for the same parameters as previously but with $F_0'^2/E_0 = 5$ and $\theta = 0.31\pi$. Clearly the motion is predominantly chaotic in both cases.
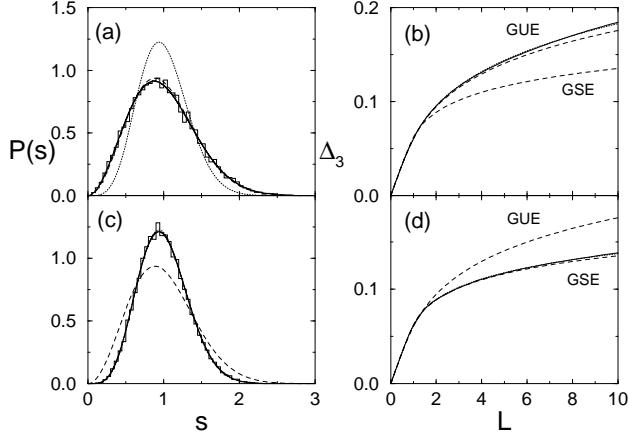
FIG. 2. Nearest neighbor spacing distribution and spectral rigidity, Δ_3 statistics, for hydrogen atom placed in a static electric field and illuminated by microwave fields, for the case with [panels (a)-(b)] and without [panels (c)-(d)] the parity symmetry $\Pi_z = \exp(i\pi S_z)P_z$ compared with the predictions of random matrices ensembles. In panels (a) and (c) solid lines indicate the best fitting Izrailev distributions, while dashed and dotted lines correspond to GUE and GSE distributions respectively (in panel (c) the dotted line is hardly visible behind the solid one). Panels (b) and (d): solid and dotted (hardly visible behind the solid lines) lines correspond to numerical data and their best fits, while dashed lines indicate GUE and GSE predictions as indicated in the figure.

Let us now consider the influence of the spin-orbit interaction in quantum case. The fine structure splitting scales as n_0^{-3}. Thus, when collecting eigenvalues for different n_0 manifolds (to improve the statistics) external fields have to be appropriately rescaled for the data to correspond to the same interesting physical situation in which external perturbations are comparable to the spin-orbit interaction. In effect we have diagonalized the quantum effective Hamiltonian for different hydrogenic manifolds in the range $n_0 = 50 - 59$ keeping constant $n_0 E_0 = 10^{-6}$, F_0^2/E_0 and $F_0'^2/E_0$ what ensures that the energy shift, with respect to the $-1/2n_0^2$ value, multiplied by n_0^3 is of the same magnitude for each n_0 manifold. For the case when H is invariant with respect to the parity symmetry, Π_z we have collected single members from each Kramers pair in the range of "$n_0^3 \times$ energy shift

with respect to $-1/2n_0^2$" between $-2 \cdot 10^{-6}$ and $3 \cdot 10^{-7}$ while for the broken symmetry case between $-3 \cdot 10^{-6}$ and -10^{-6}.
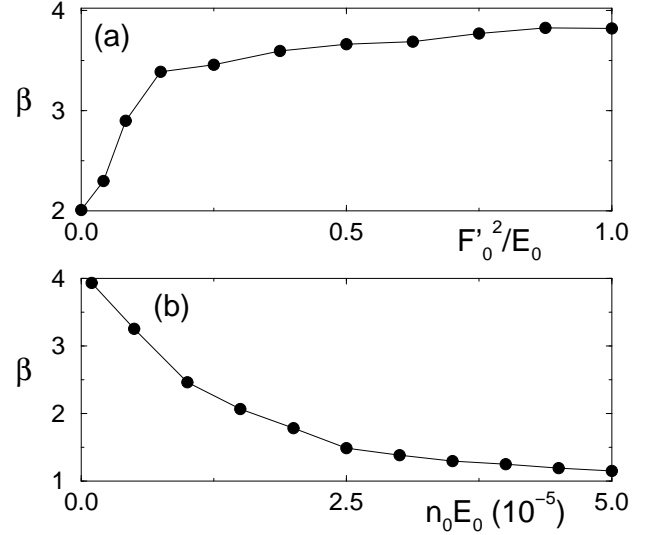
FIG. 3. The best fitting Izrailev distribution parameter, for hydrogen atom in the static electric field and driven by the two microwave fields, as a function of the parameters of the fields. Panel (a): gradual breaking of the parity symmetry, Π_z with a change of the amplitude F_0' and for fixed: $F_0^2/E_0 = 2$, $n_0 E_0 = 10^{-6}$, $\theta = \pi/2$ and $\varphi = \pi/4$. For $F_0' = 0$ the symmetry is preserved, when F_0' increases the symmetry is gradually broken and the statistics changes from the GUE to GSE type. Panel (b): *switching off* the spin-orbit interaction – keeping the configuration of the external fields fixed (i.e. $F_0^2/E_0 = 2$, $F_0'^2/E_0 = 5$, $\theta = 0.31\pi$ and $\varphi = \pi/4$) but increasing the fields amplitudes the spin-orbit interaction becomes relatively weaker. This results in a change of the level statistics from GSE to GOE type. To calculate each point in the panels the effective Hamiltonian has been diagonalized for different hydrogenic manifold in the range $n_0 = 55 - 59$.

When the Hamiltonian is invariant with respect to the Π_z transformation the nearest-neighbor spacing (NNS) distribution, $P(s)$, obtained is fitted quite satisfactorily with the distribution appropriate for GUE see Fig. 2 – top row. The solid line is a best fitting Izrailev distribution [15,16] with the parameter $\beta = 1.91$ (β gives the level repulsion exponent, i.e., $P(s) \propto s^\beta$ for small spacing s) and with $\chi^2/N = 0.4$, i.e. chi-squared divided by the number of levels N (there are about 14,000 levels in the data set). The spectral rigidities, Δ_3, also reveals the behavior very close to GUE – the best fitting Δ_3 statistics corresponding to an independent superposition of Poisson and GUE spectra [2] results in the parameter value (a relative measure of the chaotic part of the phase space) $q = 0.99$.

Breaking of the parity symmetry has a dramatic effect on the quasienergies statistics as depicted in Fig. 2 – bottom row. This time both NNS distribution and Δ_3

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statistics obtained show a very good agreement with the predictions corresponding to GSE. In particular one obtains a quartic level repulsion, $P(s) \propto s^4$ for small spacings. More precisely we have got the fitted Izrailev distribution repulsion parameter $\beta = 3.86$ with $\chi^2/N = 0.6$ and $q = 1$ from the best fitting Δ_3 statistics (now it corresponds to an independent superposition of Poisson and GSE spectra [2]). This time there are about 20,000 levels in the data set.

Finally we would like to investigate gradual breaking of the parity symmetry Π_z as well as switching off the spin-orbit interaction [2]. To this end we have diagonalized the effective Hamiltonian changing the parameters of the system. In Fig. 3a we show how the best fitting Izrailev distribution repulsion parameter, β changes when F_0' increases. That corresponds to a gradual breaking of the parity invariance – the transition from GUE type statistics to GSE one is clearly visible in the figure. *Switching off* of the spin-orbit interaction has been realized by increasing the microwave amplitudes which implies that the influence of the external fields becomes stronger while the spin-orbit interaction becomes relatively weaker. The results, as shown in Fig. 3b, indicate the transition from GSE, through intermediate, to GOE type statistics. For negligible spin-orbit interaction additional unitary symmetries are restored (e.g. $\Pi_i = \exp(i\pi S_i)$ where $i = x, y$ with the property $\Pi_i^2 = -1$) which allows splitting the Floquet Hamiltonian into two identical real matrices [3,17] and consequently the GOE statistics is expected. Actually, without the spin-orbit interaction, the spin degrees of freedom have no effect on a dynamics and can be eliminated. Then it becomes immediately clear that, because of the time-reversal invariance, the system should reveal the GOE type statistics.

To summarize, we have given an example of a realistic physical system which may yield a quartic level repulsion and, more generally, have statistical properties close to the Gaussian Symplectic Ensemble of RMT. This is possible by taking into account the spin-orbit interaction while using external perturbations (microwave and static) for creating a chaotic secular motion of the electronic ellipse. For the effect to be visible the spin-orbit and external perturbations have to be of comparable importance. This requires *very weak* external perturbation since the spin-orbit coupling decreases rapidly with n_0. The resulting mean Floquet levels spacing within the n_0 manifold is very small – on the edge of present experimental possibilities for unambiguous spectroscopy. To improve experimental conditions one may employ hydrogen-like ions since the spin-orbit interaction increases with the charge of the nucleus and consequently mean level spacing could be greater. Importantly, the Rydberg states in question have large spontaneous emission lifetimes so, at least in principle, the individual lines should be observable (and quartic level repulsion assures low probability of small spacings).

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